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Interaction of localized spins in low-temperature-grown GaAs layers

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ABSTRACT

Localized spins are associated with anti-site arsenic (As_{Ga}^{+})ions in a GaAs layer grown at a low temperature by molecular-beam epitaxy (LT-GaAs). In order to investigate possible interactions among these localized spins, magnetic properties of Be-doped LT-GaAs layers were studied by electron spin resonance (ESR) spectroscopy and super-conducting quantum interference device (SQUID) measurements, for the latter of which single crystalline LT-GaAs layers with thicknesses up to 20 μm were grown. In the SQUID measurements at strong fields, thick LT-GaAs layers show the Curie-type paramagnetism. At weak fields, a spin-glass like transition occurs; the temperature dependence of the magnetization under the zero-field cooling exhibits a peak, and the field dependence of the magnetization has a significant hysteresis at temperatures lower than the peak temperature. The peak temperature varies among samples, ranging from 2.7 to 5.5 K, depending on their growth temperatures and Be concentrations. Measured line-widths of ESR spectra have a close correlation with the results of SQUID measurements; a sample in which more significant effects of interactions among localized spins occur in the SQUID results exhibits a narrower ESR line and a larger increase in the line-width with a decrease in temperature.

1. Introduction

Localized spins in conventional magnetic materials are associated with elements containing partially filled *d* and *f* subshells. In solids, localized spins also form at defects and are normally associated with unpaired sp valence electrons. In recent years a possibility of collective magnetism based on localized spins associated with sp states has been investigated by a number of experimental and theoretical studies [1–7]. Wave functions of d and f unpaired electrons are substantially confined within magnetic ions, while sp type unpaired electrons exist in bonding or anti-bonding orbitals whose wave-functions extend to neighboring ions. As a result, magnetic interactions of localized spins associated with sp states are significantly long-ranged [7] in contrast to those between localized spins associated with d or f states. Collective magnetism of the former localized spins hence may lead to magnetic properties significantly different from those of conventional magnetic materials.

Because localized spins associated with sp states normally form at crystalline defects, it is difficult to closely control their concentration, which is a prerequisite for study on their interaction. A GaAs layer grown at a low temperature (LT-GaAs) by molecular beam epitaxy (MBE) contains a high concentration of anti-site arsenic (As_{Ga}) atoms up to $1\times10^{20}\,cm^{-3}$ [8]. The As_{Ga} center is known to be a double donor, where the (0/+) level and (+/++) level are located at E_c −0.75 eV and E_v +0.5 eV, respectively[9]. An As_{Ga}^{+} ion has an unpaired sp electron and hence possesses a localized spin that was detected by the electron spin resonance (ESR) [10] and the magnetic circular dichroism of absorption (MCDA) [11]. In non-doped LT-GaAs a few As_{Ga} atoms are ionized due to the compensation by Ga vacancies, which are deep acceptors [12]. In earlier studies on LT-GaAs, the concentration of As_{Ga}^{+} ions was found to be increased by doping a high concentration of Be atoms that compensate As_{Ga} atoms in place of Ga vacancies [13,14]. The concentration of localized spins in LT-GaAs hence can be closely controlled by doping Be atoms during the growth, while the concentration of all As_{Ga} species depends on the growth temperature for a given flux condition [15].

In the present study, we have investigated possible interactions among localized spins associated with As_{Ga}^{+} ions in LT-GaAs by increasing their concentration with the Be doping. Magnetic properties of Be-doped LT-GaAs layers were studied by ESR and super-conducting quantum interference device (SQUID) measurements, for the latter of which single crystalline LT-GaAs layers with thicknesses up to 20 μm were grown, and a substrate was removed from an LT-GaAs layer by the lift-off method. In the SQUID measurements at weak fields, a spin-glass like transition was observed from these samples. The temperature dependence of the magnetic moment under the zero-field cooling (ZFC) condition exhibited a peak, and the field dependence of the magnetic moment has a significant hysteresis at temperatures lower than the peak temperature. The peak temperature varied

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among samples, ranging from 2.7 to 5.5 K, depending on the growth temperature and the Be doping concentration. Measured line-widths of ESR spectra have a close correlation with the results of SQUID measurements; a sample in which more significant effects of interactions among localized spins occur according to the SQUID results gives rise to a narrower ESR line and a larger increase in the line-width with the decrease in temperature.

2. Experimental

Beryllium doped LT-GaAs layers were grown by utilizing a conventional MBE system. Semi-insulating epiready (1 0 0) GaAs wafers were used as substrates. After desorption of an oxide layer of the substrate surface, a 150 nm thick GaAs buffer layer was grown at 580 °C, followed by the growth of an AlAs layer and a 75 nm thick GaAs buffer layer at the same temperature. The growth of the AlAs layer whose thickness ranges from 50 to 200 nm among samples was made for the lift-off process of LT-GaAs layers [16]. The substrate temperature was subsequently lowered for the growth of an LT-GaAs layer. The growth temperature and the Be doping concentration were varied among samples. Table 1 lists samples whose spin concentrations were measured by ESR in this study. Among the listed samples, sample 1 is a non-doped LT-GaAs layer. Nearly the same atom flux ratios of As to Ga, being around 5 [15], were used for the growth of all samples. For all samples listed in Table 1, a reflection high energy electron diffraction (RHEED) pattern indicating the 2-dimensional growth mode was maintained until the end of the growth.

The Be concentrations were estimated using the Be effusion cell temperatures for which uniformly doped layers were grown at 520 °C and their hole concentrations were measured for given Be effusion cell temperatures. The substrate temperature T_s for the growth of an LT-GaAs layer was estimated by extrapolating the reading of a thermocouple at the desorption of the oxide layer on the substrate surface. It is known to be difficult to estimate accurate substrate temperatures for the low temperature MBE growth. The temperatures listed in Table 1 were not directly used for the analysis of the results in the present study because of their ambiguity.

The crystalline quality of samples was analyzed by X-ray diffraction and cross-sectional transmission electron microscopy (TEM). For the X-ray diffraction measurement, an X-ray diffractometer with a four crystal monochrometer and copper Kα radiation was used. For ESR and SQUID measurements, an LT-GaAs layer was lifted off from a GaAs substrate by etching an AlAs layer with a solution of HF acid [16]. ESR measurements were carried out with an X-band spectrometer. For the SQUID measurements 24 sheets of the LT-GaAs layer with a 3×3 mm^2 size were wrapped in a thin plastic sheet and installed in a sample holder of SQUID.

Table 1
Growth temperature, Be doping concentration, layer thickness t and free spin concentration N_s.

No.	T_s (°C)	[Be] (10^{19}/cm^3)	t (μm)	[Ns] (10^{19}/cm^3)
1	250	0	4.5	0.5
2	275	2.4	4.3	2.6
3	260	2.9	20	1.6
4	275	2.4	13	2.6
5	280	2.7	15	2.1
6	290	2.4	15	2.9

3. Results and discussion

Fig. 1 shows the ESR spectra of sample 1 and sample 2, both of which were measured at 4.5 K. The four line spectra shown in Fig. 1 are similar to that observed from LT-GaAs layers in an earlier ESR study [10], which was attributed to localized spins associated with As_{Ga}^{+} ions. A concentration of localized spins in each sample at 4.5 K was estimated with its ESR spectrum and listed in Table 1. As a reference sample for the estimation, sample 3 was used. The spin concentration in this sample at 4.5 K was derived from the measurement of the magnetic moment with SQUID at strong fields. The estimation of spin concentrations with ESR spectra was made by assuming that all samples are purely paramagnetic, but as explained later exchange interactions occur in sample 3 and other samples. The spin concentrations listed in Table 1, therefore, are considered as crudely estimated ones. A spin concentration estimated in this way for sample 1, which is a non-doped LT-GaAs layer, is close to the reported value of As_{Ga}^{+} concentration in a non-doped LT-GaAs layer grown under a similar condition [12]. As seen in Table 1, thicknesses of samples 1 and 2 are similar to each other. Fig. 1, which shows a large difference of the intensity variation between two samples, hence indicates a significant increase in the spin concentration by Be doping.

For SQUID measurements of the magnetic moment, Be-doped LT-GaAs layers with thicknesses more than 10 μm were grown. In preliminary experiments, we found it very difficult to observe magnetic moments of a sample with a thickness of only a few μm even if the spin concentration was increased by Be doping. As a number of earlier studies on the growth of LT-GaAs found, there was a breakdown of the growth of a single crystalline LT-GaAs layer at a few μm thickness or less than that thickness [17–19]. The main cause of this breakdown is surface roughening, which becomes more significant at a lower growth temperature [18,19]. In order to obtain thick LT-GaAa layers, we therefore employed relatively high growth temperatures in comparison to the normal growth temperature for LT-GaAs, which is around 200 °C. RHEED patterns indicated that the 2-dimensional growth mode was maintained until the end of the growth of all thick LT-GaAs layers. Fig. 2(a) shows an X-ray rocking curve of the (4 0 0) reflection for sample 3, whose growth was at 260 °C to a thickness of 20 μm. In the rocking curve, the 20 μm-thick LT-GaAs layer gives rise to a peak in the low angle side, while the GaAs substrate gives rise to that in the high angle side. The full-width at half maximum (FWHM) of the LT-GaAs peak, being 22 arc-sec, is comparable to

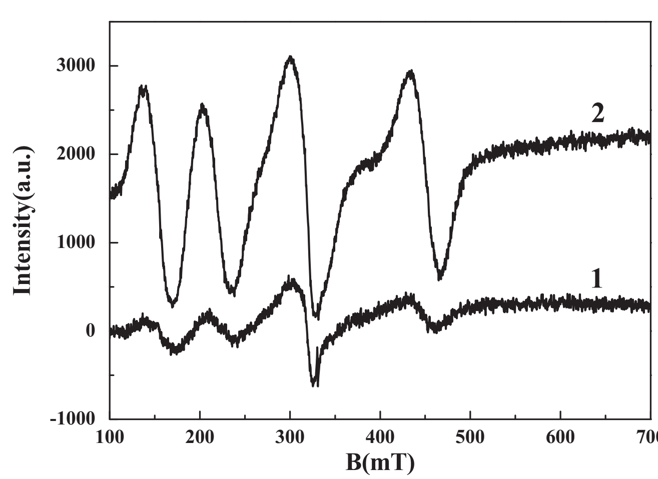

Fig. 1. ESR spectra of non-doped LT-GaAs (sample 1) and Be-doped LT-GaAs (sample 2).